van der Waals energy under strong atom-field coupling in doped carbon nanotubes

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Abstract

Using a unified macroscopic QED formalism, we derive an integral equation for the van der Waals energy of a two-level atomic system near a carbon nanotube. The equation is valid for both strong and weak atom-vacuum-field coupling. By solving it numerically, we demonstrate the inapplicability of weak-coupling-based van der Waals interaction models in a close vicinity of the nanotube surface.

Key words: A. Carbon nanotube, D. Strong atom-field coupling, D. van der Waals energy
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1 Introduction

Carbon nanotubes (CNs) are graphene sheets rolled-up into cylinders of approximately one nanometer in diameter. Extensive work carried out worldwide in recent years has revealed the intriguing physical properties of these novel molecular scale wires [1]. Nanotubes have been shown to be useful for miniaturized electronic, mechanical, electromechanical, chemical and scanning probe devices and materials for macroscopic composites [2]. Important is that their intrinsic properties may be substantially modified in a controllable way by doping with extrinsic impurity atoms, molecules and compounds [3]. Recent successful experiments on encapsulation of single atoms into single-wall CNs [4] and their intercalation into single-wall CN bundles [3,5] as well as

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the theory of spontaneous decay in such systems [6,7] stimulate an in-depth analysis of atom-CN van der Waals (vdW) interactions.

The relative density of photonic states (DOS) near a CN effectively increases due to the presence of additional surface photonic states coupled with CN electronic quasiparticle excitations [6]. This causes an atom-vacuum-field coupling constant (which is proportional to the photonic DOS) to be very sensitive to an atom-CN-surface distance. In particular [7], when the atom is close enough to the CN surface and the atomic transition frequency is in the vicinity of the resonance of the photonic DOS, the system shows *strong* atom-vacuum-field coupling giving rise to rearrangement ("dressing") of atomic levels by vacuum-field interaction. If the atom moves away from the CN surface, the atom-field coupling strength decreases, smoothly approaching the weak coupling regime at large atom-surface distances since the role of surface photonic states diminishes causing the relative density of photonic states to decrease. This suggests strictly *nonlinear* atom-field coupling and a *primary* role of the distance-dependent (local) photonic DOS in the vicinity of the CN, so that vacuum-QED-based vdW interaction models as well as those based upon the linear response theory (see [8] for a review) are in general inapplicable for an atom in a close vicinity of a carbon nanotube.

To give this issue a proper consideration, we have developed a simple quantum mechanical approach to the vdW energy calculation of a (two-level) atomic system in the vicinity of an infinitely long single-wall CN. The approach is based upon the perturbation theory for degenerate atomic levels (see, e.g., [9]), thereby allowing one to account for both strong and weak atom-vacuum-field coupling regimes. In describing the atom-field interaction, we follow a unified macroscopic QED formalism developed for dispersing and absorbing media in Ref. [10] and adapted for an atom near a CN in Refs. [6,7]. The formalism generalizes the standard macroscopic QED normal-mode technique by representing mode expansion in terms of a Green tensor of the (operator) Maxwell equations in which material dispersion and absorbtion are automatically included. In more detail, the Fourier-images of electric and magnetic fields are considered as quantum mechanical observables of corresponding electric and magnetic field operators. The latter ones satisfy the Fourier-domain operator Maxwell equations modified by the presence of a so-called operator noise current written in terms of a bosonic field operator and an imaginary dielectric permittivity. This operator is responsible for correct commutation relations of the electric and magnetic field operators in the presence of medium-induced absorbtion. The electric and magnetic field operators are then expressed in terms of a continuum set of operator bosonic fields by means of the convolution of the operator noise current with the electromagnetic field Green tensor of the system. The bosonic field operators create and annihilate single-quantum electromagnetic medium excitations. They are defined by their commutation relations and play the role of the fundamental dynamical variables in terms

2

of which the Hamiltonian of the composed system "electromagnetic field + dissipative medium" is written in a standard secondly quantized form.

Using the approach summarized above, we derive an integral equation for the vdW energy of a two-level atomic system near a CN. The equation is represented in terms of the local photonic DOS and is valid for both strong and weak atom-field coupling. By solving it numerically, we demonstrate the inapplicability of weak-coupling-based vdW interaction models in a close vicinity of the CN.

2 The van der Waals energy of an atom near a carbon nanotube

Consider a neutral atomic system with its centre-of-mass positioned at the point $\mathbf{r}_A$ near an infinitely long single-wall CN. Assign the orthonormal cylindric basis $\{\mathbf{e}_r, \mathbf{e}_\varphi, \mathbf{e}_z\}$ with $\mathbf{e}_z$ directed along the CN axis. The total nonrelativistic Hamiltonian of the whole system can then be represented in the form (electric-dipole approximation, Coulomb gauge, CGS units) [7,8]

$$\hat{H} = \hat{H}_F + \hat{H}_A + \hat{H}_{AF}^{(1)} + \hat{H}_{AF}^{(2)}, \tag{1}$$

where

$$\hat{H}_F = \int\limits_0^\infty d\omega \hbar\omega \int d\mathbf{R} \, \hat{f}^\dagger(\mathbf{R}, \omega) \hat{f}(\mathbf{R}, \omega), \tag{2}$$

$$\hat{H}_A = \sum_i \frac{\hat{\mathbf{p}}^2}{2m_i} + \frac{1}{2} \sum_{i,j} \frac{q_i q_j}{|\mathbf{r}_i - \mathbf{r}_j|}, \tag{3}$$

$$\hat{H}_{AF}^{(1)} = -\sum_i \frac{q_i}{m_i c} \hat{\mathbf{p}}_i \cdot \hat{\mathbf{A}}(\mathbf{r}_A) + \hat{\mathbf{d}} \cdot \nabla \hat{\varphi}(\mathbf{r}_A), \tag{4}$$

$$\hat{H}_{AF}^{(2)} = \sum_i \frac{q_i^2}{2m_i c^2} \hat{\mathbf{A}}^2(\mathbf{r}_A) \tag{5}$$

are, respectively, the Hamiltonian of the vacuum electromagnetic field modified by the presence of the CN, the Hamiltonian of the atomic subsystem, and the Hamiltonian of their interaction (separated into two contributions according to their role in the atom-CN vdW interaction – see below). The operators $\hat{f}^\dagger$ and $\hat{f}$ in Eq. (2) are those creating and annihilating single-quantum electromagnetic excitations of bosonic type in the CN and the inner integral is taken over the CN surface assigned by the vector $\mathbf{R} = \{R_{cn}, \phi, Z\}$ with R_{cn} being the CN radius. In Eqs. (3)-(5), m_i, q_i, $\hat{\mathbf{r}}_i$ and $\hat{\mathbf{p}}_i$ are, respectively, the masses,

charges, canonically conjugated coordinates (relative to $\mathbf{r}_A$) and momenta of the particles constituting the atomic subsystem, $\hat{\mathbf{d}} = \sum_i q_i \hat{\mathbf{r}}_i$ is its electric dipole moment operator. The vector potential $\hat{\mathbf{A}}$ and the scalar potential $\hat{\varphi}$ of the CN-modified electromagnetic field are given for an arbitrary $\mathbf{r} = \{r, \varphi, z\}$ in the Schrödinger picture by

$$\hat{\mathbf{A}}(\mathbf{r}) = \int\limits_0^\infty d\omega (i\omega)^{-1} \underline{\hat{\mathbf{E}}}^\perp(\mathbf{r}, \omega) + \text{h.c.}, \quad -\nabla\hat{\varphi}(\mathbf{r}) = \int\limits_0^\infty d\omega\, \underline{\hat{\mathbf{E}}}^\parallel(\mathbf{r}, \omega) + \text{h.c.}, \quad (6)$$

where $\underline{\hat{\mathbf{E}}}^{\perp(\parallel)}(\mathbf{r}, \omega) = \int d\mathbf{r}'\, \boldsymbol{\delta}^{\perp(\parallel)}(\mathbf{r} - \mathbf{r}') \cdot \hat{\mathbf{E}}(\mathbf{r}', \omega)$ is the transverse (longitudinal) electric field with $\delta_{\alpha\beta}^\parallel(\mathbf{r}) = -\nabla_\alpha \nabla_\beta (4\pi r)^{-1}$ and $\delta_{\alpha\beta}^\perp(\mathbf{r}) = \delta_{\alpha\beta}\delta(\mathbf{r}) - \delta_{\alpha\beta}^\parallel(\mathbf{r})$ being the longitudinal and transverse dyadic δ-functions, respectively, and $\underline{\hat{\mathbf{E}}}$ representing the total electric field operator which satisfies the following set of Fourier-domain Maxwell equations

$$\nabla \times \underline{\hat{\mathbf{E}}}(\mathbf{r}, \omega) = ik\, \underline{\hat{\mathbf{H}}}(\mathbf{r}, \omega), \quad (7)$$

$$\nabla \times \underline{\hat{\mathbf{H}}}(\mathbf{r}, \omega) = -ik\, \underline{\hat{\mathbf{E}}}(\mathbf{r}, \omega) + \frac{4\pi}{c}\underline{\hat{\mathbf{I}}}(\mathbf{r}, \omega).$$

Here, $\underline{\hat{\mathbf{H}}}$ stands for the magnetic field operator, $k = \omega/c$, and

$$\underline{\hat{\mathbf{I}}}(\mathbf{r}, \omega) = \int d\mathbf{R}\, \delta(\mathbf{r} - \mathbf{R})\, \underline{\hat{\mathbf{J}}}(\mathbf{R}, \omega) = 2\underline{\hat{\mathbf{J}}}(R_{cn}, \varphi, z, \omega)\, \delta(r - R_{cn}), \quad (8)$$

where

$$\underline{\hat{\mathbf{J}}}(\mathbf{R}, \omega) = \sqrt{\frac{\hbar\omega \mathrm{Re}\sigma_{zz}(\mathbf{R}, \omega)}{\pi}}\, \hat{f}(\mathbf{R}, \omega)\, \mathbf{e}_z \quad (9)$$

is the operator noise current density associated with CN-induced absorbtion [7], σ_{zz} is the CN surface axial conductivity per unit length (in describing CN electronic properties, we use the surface axial conductivity model, neglecting the CN azimuthal current and radial polarizability [11]).

From Eqs. (7)-(9) it follows that

$$\underline{\hat{\mathbf{E}}}(\mathbf{r}, \omega) = i\frac{4\pi}{c}\, k \int d\mathbf{R}\, \mathbf{G}(\mathbf{r}, \mathbf{R}, \omega) \cdot \underline{\hat{\mathbf{J}}}(\mathbf{R}, \omega) \quad (10)$$

(and $\underline{\hat{\mathbf{H}}} = (ik)^{-1}\nabla \times \underline{\hat{\mathbf{E}}}$ accordingly), where $\mathbf{G}$ is the Green tensor of the classical electromagnetic field in the vicinity of the CN. The set of Eqs. (1)-(10) forms a closed electromagnetic field quantization formalism in the presence

of dispersing and absorbing media which meets all the basic requirements of a standard quantum electrodynamics [10]. All information about medium (the CN in our case) is contained in the Green tensor $\mathbf{G}$ whose components satisfy the equation

$$\sum_{\alpha=r,\varphi,z} \left(\nabla \times \nabla \times - k^2 \right)_{z\alpha} G_{\alpha z}(\mathbf{r}, \mathbf{R}, \omega) = \delta(\mathbf{r} - \mathbf{R}), \tag{11}$$

together with the radiation conditions at infinity and the boundary conditions on the CN surface. This tensor was derived and analysed in Ref. [7].

Starting from Eqs. (1)-(5) and using Eqs. (6), (9), (10), one obtains the following secondly quantized Hamiltonian for a *two-level* atomic system interacting with the CN-modified electromagnetic field

$$\hat{H} = \int_0^\infty d\omega \hbar\omega \int d\mathbf{R} \, \hat{f}^\dagger(\mathbf{R}, \omega) \hat{f}(\mathbf{R}, \omega) + \frac{\hbar\tilde{\omega}_A}{2} \, \hat{\sigma}_z \tag{12}$$

$$+ \int_0^\infty d\omega \int d\mathbf{R} \left[\mathrm{g}^{(+)}(\mathbf{r}_A, \mathbf{R}, \omega) \, \hat{\sigma}^\dagger - \mathrm{g}^{(-)}(\mathbf{r}_A, \mathbf{R}, \omega) \, \hat{\sigma} \right] \hat{f}(\mathbf{R}, \omega) + \mathrm{h.c.}.$$

Here, the Pauli operators $\hat{\sigma}_z = |u\rangle\langle u| - |l\rangle\langle l|$, $\hat{\sigma} = |l\rangle\langle u|$, $\hat{\sigma}^\dagger = |u\rangle\langle l|$ describe electric dipole transitions between the two atomic states, upper $|u\rangle$ and lower $|l\rangle$, separated by the transition frequency ω_A. This (bare) frequency is modified by the interaction (5) which, being independent of the atomic dipole moment, does not contribute to mixing the $|u\rangle$ and $|l\rangle$ states, giving rise, however, to the new *renormalized* transition frequency

$$\tilde{\omega}_A = \omega_A \left(1 - \frac{2}{(\hbar\omega_A)^2} \int_0^\infty d\omega \int d\mathbf{R} \, |\mathrm{g}^\perp(\mathbf{r}_A, \mathbf{R}, \omega)|^2 \right) \tag{13}$$

in the second term of Eq. (12). On the contrary, the interaction (4), being dipole moment dependent, mixes the $|u\rangle$ and $|l\rangle$ states, yielding the third term of the Hamiltonian (12) with the interaction matrix elements

$$\mathrm{g}^{(\pm)}(\mathbf{r}_A, \mathbf{R}, \omega) = \mathrm{g}^\perp(\mathbf{r}_A, \mathbf{R}, \omega) \pm \frac{\omega}{\omega_A} \mathrm{g}^\parallel(\mathbf{r}_A, \mathbf{R}, \omega), \tag{14}$$

where

$$\mathrm{g}^{\perp(\parallel)}(\mathbf{r}_A, \mathbf{R}, \omega) = -i\frac{4\omega_A}{c^2} \sqrt{\pi\hbar\omega \mathrm{Re}\sigma_{zz}(\omega)} \, d_\alpha G_{\alpha z}^{\perp(\parallel)}(\mathbf{r}_A, \mathbf{R}, \omega) \tag{15}$$

with $G_{\alpha z}^{\perp(\|)}(\mathbf{r}_A, \mathbf{R}, \omega) = \int d\mathbf{r}\, \delta_{\alpha\beta}^{\perp(\|)}(\mathbf{r}_A - \mathbf{r})\, G_{\beta z}^{\perp(\|)}(\mathbf{r}, \mathbf{R}, \omega)$. The matrix element (15), being squared and integrated over the nanotube surface, may be represented in the form

$$\int d\mathbf{R}\, |\mathrm{g}^{\perp(\|)}(\mathbf{r}_A, \mathbf{R}, \omega)|^2 = \hbar^2 \frac{\Gamma_0(\omega)}{2\pi} \left(\frac{\omega_A}{\omega}\right)^2 \xi^{\perp(\|)}(\mathbf{r}_A, \omega), \tag{16}$$

where $\xi^{\perp(\|)}$ is the transverse (longitudinal) *local* photonic DOS defined by [7]

$$\xi^{\perp(\|)}(\mathbf{r}_A, \omega) = \frac{\Gamma^{\perp(\|)}(\mathbf{r}_A, \omega)}{\Gamma_0(\omega)} \tag{17}$$

with $\Gamma^{\perp(\|)}(\mathbf{r}_A, \omega) = 8\pi d_\alpha d_\beta \,\mathrm{Im}^{\perp(\|)} G_{\alpha\beta}^{\perp(\|)}(\mathbf{r}_A, \mathbf{r}_A, \omega)/\hbar c^2$ being the transverse (longitudinal) atomic spontaneous decay rate near the CN, and Γ_0 representing the same quantity in vacuum where $\mathrm{Im} G_{\alpha\beta}^0 = \delta_{\alpha\beta}\, \omega/6\pi c$. Note that $\mathrm{Im}^{\perp(\|)} G_{\alpha\beta}^{\perp(\|)} = \mathrm{Im} G_{\alpha\beta}^0 + \mathrm{Im}^{\perp(\|)}\overline{G}_{\alpha\beta}^{\perp(\|)}$ with the second term representing the "pure" CN contribution to the total imaginary Green tensor, so that Eq. (17) may also be written in the form $\xi^{\perp(\|)}(\mathbf{r}_A, \omega) = 1 + \overline{\xi}^{\perp(\|)}(\mathbf{r}_A, \omega)$.

The Hamiltonian (12) is a starting point for the vdW energy calculation. The latter is nothing but the $\mathbf{r}_A$-dependent contribution to the ground-state energy given by this Hamiltonian. Important is that the two-level atomic subsystem (the second term in Eq. (12)) is now described by the *renormalized* frequency (13) which is seen (see Eq. (16)) to decrease with increasing $\xi^\perp$ (i.e. when the atom approaches the CN surface [6,7]), thereby bringing the two atomic levels together, or even making them degenerated if $\xi^\perp$ is large enough. To take this fact into account in a correct way, one has to calculate the energy using the perturbation theory for degenerated levels (see, e.g., [9]). In so doing, one also has to account for the upper state degeneracy of the whole system with respect to $\mathbf{R}$ and ω. In view of this, the wave function of the whole system should be written in the form

$$|\psi\rangle = C_l\, |l\rangle|\{0\}\rangle + \int\limits_0^\infty d\omega \int d\mathbf{R}\, C_u(\mathbf{R}, \omega)\, |u\rangle|\{1(\mathbf{R}, \omega)\}\rangle, \tag{18}$$

where $|\{0\}\rangle$ is the vacuum state of the field subsystem, $|\{1(\mathbf{R}, \omega)\}\rangle$ is its single-quantum excited state, C_l and C_u are unknown mixing coefficients of the lower and upper states of the *whole* system. The total ground-state energy E is then given by the solution of a secular equation obtained by applying the Hamiltonian (12) to the wave function (18). This yields the integral equation

$$E = -\frac{\hbar\tilde{\omega}_A}{2} - \int\limits_0^\infty d\omega \int d\mathbf{R}\, \frac{|\mathrm{g}^{(-)}(\mathbf{r}_A, \mathbf{R}, \omega)|^2}{\hbar\omega + \dfrac{\hbar\tilde{\omega}_A}{2} - E}, \tag{19}$$

which the vdW energy E_{vw} is determined from by means of the obvious relation $E = -\hbar\omega_A/2 + E_{vw}(\mathbf{r}_A)$ where the first term is the unperturbed ground-state energy of the two-level atom. Using further the dimensionless variables $\varepsilon_{vw} = E_{vw}/2\gamma_0$ and $x = \hbar\omega/2\gamma_0$ ($\gamma_0 = 2.7$ eV is the carbon nearest neighbor hopping integral appearing in σ_{zz} in Eq. (9)), one finally arrives, in view of Eqs. (13)-(16), at the following integral equation

$$\varepsilon_{vw}(\mathbf{r}_A) = \frac{\hbar}{3\pi\gamma_0 x_A}\int\limits_0^\infty dx \Gamma_0(x)\overline{\xi}^\perp(\mathbf{r}_A, x) \tag{20}$$

$$-\frac{\hbar}{3\pi\gamma_0}\int\limits_0^\infty dx \frac{\Gamma_0(x)\left[\overline{\xi}^\perp(\mathbf{r}_A, x) + \left(\frac{x}{x_A}\right)^2\overline{\xi}^\parallel(\mathbf{r}_A, x)\right]}{x + x_A - \dfrac{\hbar}{3\pi\gamma_0 x_A}\int\limits_0^\infty dx \Gamma_0(x)\overline{\xi}^\perp(\mathbf{r}_A, x) - \varepsilon_{vw}(\mathbf{r}_A)}$$

describing the vdW energy in terms of the distance-dependent photonic DOS analyzed in great detail in Ref. [7] and shown in Figure 1(a) for a particular case of the atom outside the (9,0) CN. Eq. (20) is *universal* in the sense that it covers both strong and weak atom-field coupling regimes. These two limiting cases are obtained by putting $x_A \approx (\hbar/3\pi\gamma_0 x_A)\int_0^\infty dx\Gamma_0(x)\overline{\xi}^\perp(\mathbf{r}_A, x)$ and $\varepsilon_{vw} \approx 0$, respectively, in the denominator of the second term. The former is nothing but a degeneracy condition of the unperturbed atomic levels due to the atom-field interaction, taking place at small atom-CN-surface distances when the local photonic DOS is large enough. The latter occurs at lower photonic DOS values when the atom is not very close to the CN surface. In this latter case, Eq. (20) gives rise to an ordinary second order perturbation theory result with the total Hamiltonian (12) where the last term is considered as a perturbation. This result is slightly "improved" compared with those obtained from various weak-coupling-based models discussed in the literature (see [8] and Refs. therein) since a part of the atom-field interaction is included in the unperturbed Hamiltonian via the renormalized transition frequency.

3 Numerical results and discussion

Using Eq. (20), we have simulated the vdW energy of the atom outside the (9,0) CN. The local photonic DOS functions $\overline{\xi}^{\perp,\parallel}(\mathbf{r}_A, x) = \xi^{\perp,\parallel}(\mathbf{r}_A, x) - 1$ were computed in the same manner as it was done in Ref. [7]. The free-space spontaneous decay rate was approximated by the expression $\Gamma_0(x) \approx \alpha^3 2\gamma_0 x/\hbar$ ($\alpha = 1/137$ is the fine-structure constant) valid for hydrogen-like atoms [9]. Figure 1(a) shows the transverse local photonic DOS, as a typical example, for the atom located at several distances from the CN surface. The DOS function
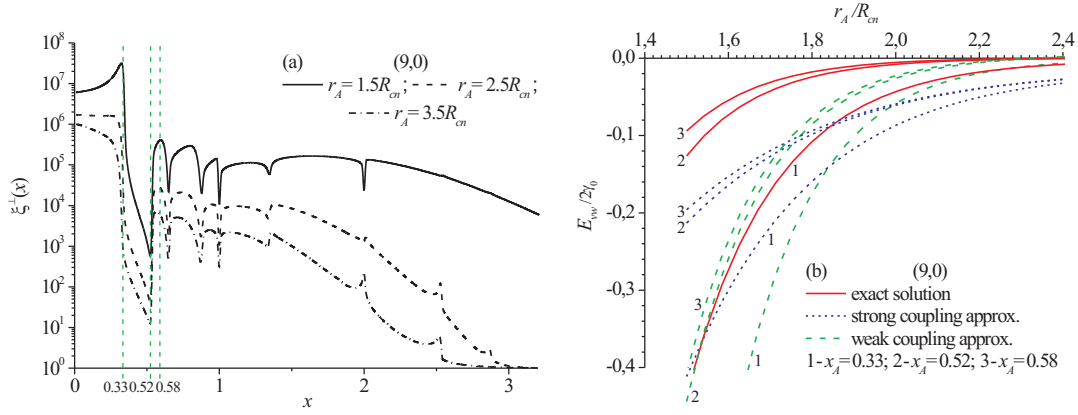
Fig. 1. Typical example of photonic DOS (the transverse local photonic DOS) (a) and the vdW energy (b) of the atom outside the (9,0) CN. The (bare) atomic transition frequencies are indicated by dashed lines in Fig. 1(a).

is seen to increase with decreasing the atom-CN-surface distance, representing the increase of the atom-field coupling strength as the atom approaches the nanotube surface [7]. The vertical dashed lines indicate the bare atomic transitions frequencies x_A for which the vdW energies shown in Figure 1(b) were calculated. The frequencies are typical for heavy hydrogen-like atoms such as Cs for which x_A may be estimated from its first ionization potential [12] to be $3.89\,\text{eV} \times 3/4 \times (2\gamma_0)^{-1} \sim 0.5$ (the factor 3/4 comes from the Lyman series of H), or less for highly excited Rydberg states.

In Figure 1(b), the vdW energies given by exact numerical solutions of Eq. (20) are compared with those obtained from the same equation within the weak and strong coupling approximations. At small atom-CN-surface distances, the exact solutions are seen to be fairly well reproduced by those obtained in the strong coupling approximation, clearly indicating the strong atom-vacuum-field coupling regime in a close vicinity of the nanotube surface. The deviation from the strong coupling approximation increases with x_A, that is easily explicable since the degeneracy condition of the unperturbed atomic levels is more difficult to reach for larger interlevel separations. As the atom moves away from the CN surface, the exact solutions deviate from the strong coupling solutions and approach those given by the weak coupling approximation, indicating the reduction of the atom-field coupling strength with raising the atom-surface distance. The weak-coupling solutions are seen to be divergent close to the nanotube surface as it should be because of the degeneracy of the unperturbed (bare) atomic levels in this region.

To conclude, we have investigated vdW interactions of a two-level atomic system near a single-wall CN within the unified QED approach accounting for both strong and weak atom-vacuum-field coupling. We have demonstrated the

inapplicability of weak-coupling-based vdW interaction models in a close vicinity of the nanotube surface where the local photonic DOS effectively increases giving rise to atom-field coupling enhancement followed by the degeneracy and rearrangement ("dressing") of bare atomic levels.

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